

Spin dynamics of a tetrahedral cluster magnet

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We study the magnetism of a lattice of coupled tetrahedral spin-1/2 clusters which might be of relevance to the tellurate compounds $\text{Cu}_2\text{Te}_2\text{O}_5\text{X}_2$, with $\text{X}=\text{Cl}, \text{Br}$. Using the flow equation method we perform a series expansion in terms of the inter-tetrahedral exchange couplings starting from the quadrumer limit. Results will be given for the magnetic instabilities of the quadrumer phase and the dispersion of elementary triplet excitations. In limiting cases of our model of one- or two dimensional character we show our results to be consistent with findings on previously investigated decoupled tetrahedral chains and the Heisenberg model on the 1/5-depleted square lattice.

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I. INTRODUCTION

Unconventional magnetism of frustrated spin systems has received considerable interest recently. Prominent examples are the one-dimensional (1D) frustrated spin-Peierls compound CuGeO_3 ¹ or the 2D orthogonal spin-dimer system $\text{SrCu}_2(\text{BO}_3)_2$ with frustrating inter-dimer couplings². Apart from dimer-based structures, frustrated spin systems involving triangular or tetrahedral units, e.g. the kagomé-, checkerboard-, or the pyrochlore-lattices are a focus of current research. In the classical limit frustration leads to ground states with macroscopic degeneracy in these systems^{3,4}. In the quantum case low-lying singlets seem to exist both, on the kagomé and the checkerboard lattice with no long-range magnetic order in the former^{4,5}, and a valence-bond-crystal (VBC) ground state in the latter case^{6,7}. Analysis of the 3D pyrochlore quantum-magnet remains an open issue⁸.

Recently tellurate compounds, $\text{Cu}_2\text{Te}_2\text{O}_5\text{X}_2$, with $\text{X} = \text{Cl}, \text{Br}$ have been found to realize a new class of spin-1/2 systems where tetrahedra of Cu^{2+} align in tubes along the $c(z)$ -direction and are separated by lone-pair cations in the $ab(xy)$ -plane⁹. Both, the effective dimensionality of this system as well as the relevant magnetic interactions remain a puzzle. Early analysis of thermodynamic data^{9,10} was based on the 0D limit of isolated

tetrahedral units, i.e. on the exchange pattern of fig. 1 with $j_{2,\dots,6} = 0$. This resulted in $j_0 = 38.5(43)\text{K}$ and $j_1/j_0 \sim 1$ for the Chlorine(Bromide) system which has been refined recently into $j_0 \approx 47.66\text{K}$ and $j_1/j_0 \approx 0.66$ for the Bromide system and $j_1/j_0 < 0.66$ for the Chlorine case¹¹. Raman spectroscopy¹⁰ however, indicates a substantial inter-tetrahedral c -axis coupling. This has prompted studies of 1D tetrahedral spin-chains^{12,13} as in fig. 1 with $j_{2,3,4,6} = 0$. Yet, LDA calculations have given evidence of an additional z -axis exchange path j_6 and transverse inter-chain couplings as shown in 1 a) of a magnitude which can not be neglected¹⁰. In fact, specific heat data reveals a transition at $T_C = 18.2(11.4)\text{K}$ in the Chlorine(Bromide) system. In the Chlorine case the entropy change is consistent with 3D antiferromagnetic (AFM) ordering.

Combining fig. 1 a) and b) a 3D cluster-spin model arises, about which very little is known. We believe this to be an interesting and highly frustrated magnetic system which deserves to be investigated. Therefore, the aim of this work is to shed light onto its excitations and possible magnetic instabilities. In addition, our analysis could be of relevance for the $\text{Cu}_2\text{Te}_2\text{O}_5\text{X}_2$ system, in particular, if additional spectroscopic data becomes available. In the remainder of this paper we first discuss our method of calculation and then present results on the stability and the triplet dynamics.

II. SERIES EXPANSION

The Hamiltonian, as read off from fig. 1 can split into a bare part H_0 and a perturbation H_1

$$\begin{aligned} H &= H_0 + H_1 \\ H_0 &= \sum_i j_0 (\mathbf{S}_{1i} + \mathbf{S}_{3i})(\mathbf{S}_{2i} + \mathbf{S}_{4i}) \\ H_1 &= \sum_i [j_1 (\mathbf{S}_{1i}\mathbf{S}_{3i} + \mathbf{S}_{2i}\mathbf{S}_{4i}) \\ &\quad + j_2 (\mathbf{S}_{4i}\mathbf{S}_{1i+x} + \mathbf{S}_{3i}\mathbf{S}_{2i+x} \\ &\quad + \mathbf{S}_{2i}\mathbf{S}_{1i+y} + \mathbf{S}_{3i}\mathbf{S}_{4i+y}) + \end{aligned}$$

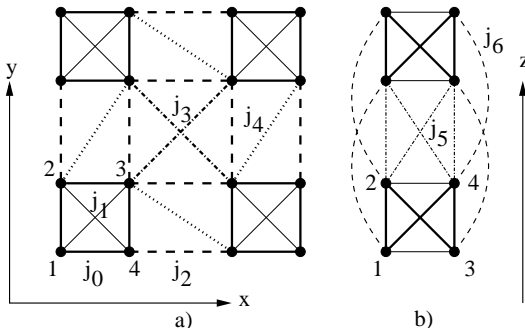


FIG. 1: a) xy -plane and b) z -axis structure of the 3D tetrahedral cluster-lattice. Spin-1/2 moments are located on dotted vertices. $SU(2)$ type of exchange with strength $j_{0,\dots,6}$ along the links.

$$\begin{aligned}
& +j_3(\mathbf{S}_{3l}\mathbf{S}_{1l+\mathbf{x}+\mathbf{y}} + \mathbf{S}_{2l}\mathbf{S}_{4l-\mathbf{x}+\mathbf{y}}) \\
& +j_4(\mathbf{S}_{3l}\mathbf{S}_{1l+\mathbf{x}} + \mathbf{S}_{2l}\mathbf{S}_{4l+\mathbf{y}}) \\
& +j_5(\mathbf{S}_{2l} + \mathbf{S}_{4l})(\mathbf{S}_{1l+\mathbf{z}} + \mathbf{S}_{3l+\mathbf{z}}) \\
& +j_6 \sum_i \mathbf{S}_{il}\mathbf{S}_{i+\mathbf{z}} \quad , \quad (1)
\end{aligned}$$

where the site of each tetrahedral unit is labeled by \mathbf{l} with \mathbf{S}_{il} , $i = 1 \dots 4$ being the spin-1/2 operators corresponding to each tetrahedron and $\mathbf{l} + \mathbf{x}(\mathbf{y}, \mathbf{z})$ refers to shifts of \mathbf{l} by one unit cell along the x , y , or z -axis.

As has been pointed out previously⁹ the spectrum of decoupled tetrahedra, i.e. for $j_{2,\dots,6} = 0$, is special in as such that for $j_1 < j_0$ each tetrahedral ground state is a singlet involving all four spins, while at $j_1 > j_0$ it is a product of two $S=0$ -dimers on each of the j_1 -bonds. In turn, at $j_1 = j_0$ the decoupled local ground states are doubly degenerate singlets. This leads to quantum criticality in the lattice case and bears the possibility of low-lying singlet excitations^{9,12,13}. This may be relevant for the Bromide system but in the Chlorine compound j_1 seems clearly less than j_0 . Therefore in the remainder of this paper we focus on the *quadrumer limit* of (1), defined by setting $j_0 \equiv 1$ and $j_{1,\dots,6} \ll 1$.

The spectrum of each quadrumer consists of four *equidistant* levels which can be labeled by spin S and a number of local energy quanta q_1 , c.f. table I. The unperturbed Hamiltonian H_0 which consists of the sum of quadrumers displays an equidistant ladder-spectrum labeled by $Q = \sum_1 q_1$. The $Q = 0$ sector is the unperturbed ground state $|0\rangle$ of H_0 , which is a VBC of quadrumer-singlets. The $Q = 1$ -sector contains local $S = 1$ single-particle excitations of the VBC with $q_1 = 1$, where \mathbf{l} runs over the lattice. At $Q = 2$ the spectrum of H_0 has total $S = 0, 1$, or 2 and is of multi-particle nature. For $S = 0$ at $Q = 2$ it comprises of one-particle singlets with $q_1 = 2$ and two-particle singlets constructed from triplets with $q_1 = q_{\mathbf{m}} = 1$ and $\mathbf{l} \neq \mathbf{m}$. The perturbation H_1 in (1) can be written as a sum of *two-site* operators $T_{n,k}$ which, for each coupling constant $j_{k=1,\dots,6}$ create(destroy) $n \geq 0$ ($n < 0$) quanta within the ladder spectrum of H_0 .

$$H = H_0 + \sum_{n=-N}^N \sum_{k=1}^6 j_k T_{n,k} \quad (2)$$

It has been shown recently^{7,14,15,16} that problems of type (2) allow for perturbative analysis using a continuous unitary transformation generated by the flow equation method of Wegner¹⁷. The unitarily rotated effective Hamiltonian H_{eff} reads^{14,16}

$$H_{\text{eff}} = H_0 + \sum_{n=1}^{\infty} \sum_{\substack{|\mathbf{m}|=n \\ M(\mathbf{m})=0}} C(\mathbf{m}) W_{m_1} W_{m_2} \dots W_{m_n} \quad (3)$$

where $\mathbf{m} = (m_1 \dots m_n)$ with $|\mathbf{m}| = n$ is an n -tuple of integers, each in a range of $m_i \in \{0, \pm 1, \dots, \pm N\}$ and

E	S	q_1
1	2	3
0	$0 \oplus 1 \oplus 1$	2
-1	1	1
-2	0	0

TABLE I: Energy (E , in units of j_0), spin (S), and quantum-number q_1 of the quadrumer spectrum.

$W_n = \sum_{k=1}^6 j_k T_{n,k}$. In contrast to H of (1), H_{eff} *conserves* the total number of quanta Q . This is evident from the constraint $M(\mathbf{m}) = \sum_{i=1}^n m_i = 0$. The amplitudes $C(\mathbf{m})$ are rational numbers computed from the flow equation method^{14,16}. Explicit tabulation¹⁸ of the $T_{n,k}$ shows that for the Hamiltonian in (1) $N = 4$.

Q -conservation of H_{eff} leads to a ground state energy of $E_g = \langle 0 | H_{\text{eff}} | 0 \rangle$. Evaluating this matrix element on clusters with periodic boundary conditions, sufficiently large not to allow for wrap around at graph-length n one can obtain series expansions (SEs) for E_g valid to $O(n)$ in the thermodynamic limit, i.e. for systems of infinite size. Q -conservation also guarantees the $Q = 1$ -triplets remain genuine one-particle states. *A priori* single-particle states from sectors with $Q > 1$ will not only disperse via H_{eff} , but can decay into multi-particle states. The dispersion of the single-particle excitations is

$$E_{\mu}(\mathbf{k}) = \sum_{lm} t_{\mu,lm} e^{i(k_x l + k_y m)} \quad (4)$$

where $t_{\mu,lm} = \langle \mu, lm | H_{\text{eff}} | \mu, 00 \rangle - \delta_{lm,00} E_g^{obc}$ are hopping matrix elements from site $(0,0)$ to site (l,m) for a quadrumer excitation μ inserted into the unperturbed ground state. For the thermodynamic limit $t_{\mu,lm}$ has to be evaluated on clusters with open boundary conditions large enough to embed all linked paths of length n connecting sites $(0,0)$ to (l,m) at $O(n)$ of the perturbation. $E_g^{obc} = \langle 0 | H_{\text{eff}} | 0 \rangle$ on the $t_{\mu,00}$ -cluster.

Previous applications of this method to spin systems were focused on obtaining high-order SEs for one and two parameter dimer¹⁶ and quadrumer⁷ models in 1 or 2D. In the present case, computational constraints related to the large number of coupling constants and the 3D nature of the model confine the expansion to 4th-order. Moreover, explicit display of analytic expressions for the elementary triplet dispersion has to be limited to 2nd order¹⁸.

III. TRIPLET EXCITATIONS AND MAGNETIC INSTABILITIES

In this section we analyze the triplet dispersion $E_T(\mathbf{k})$ and the stability of the quadrumer phase against magnetic ordering. We begin by considering the result at 1st

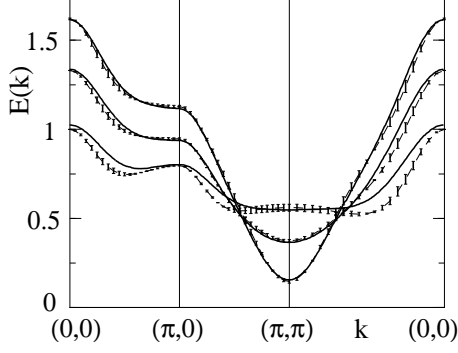


FIG. 2: Comparing the triplet dispersion along high-symmetry directions of a 2D Brillouin zone as obtained from a 6th-order plaquette expansion for the 1/5-depleted square lattice²¹ (dashed with error bars) with the 4th-order quadrumer expansion for the tetrahedral spin-cluster model (solid) at $j_1 = \gamma$, $j_2 = \lambda\gamma$, $j_4 = \lambda$, and $j_{3,5,6} = 0$, with $\lambda = 1$ and $\gamma = 0.1, 0.3$, and 0.5 from top to bottom. (Upper and lower edges of error bars refer to 4th- and 5th-order plaquette expansion.)

order in $j_{1,\dots,6}$ for which we find

$$E_T(\mathbf{k}) = 1 + \frac{1}{3}(j_4 - 2j_2)(\cos(k_x) + \cos(k_y)) + \frac{j_3}{3}(\cos(k_x + k_y) + \cos(k_x - k_y)) + \frac{4}{3}(j_6 - j_5)\cos(k_z). \quad (5)$$

Interestingly, this expression depends on three, effective exchange coupling-constants only, i.e. $a = (j_4 - 2j_2)/3$, $b = j_3/3$, and $c = 4(j_6 - j_5)/3$. Moreover $E_{\mathbf{k}}$ is independent of j_1 . In fact we find the dispersion to depend on j_1 starting only at 3rd order. Due to the competition of the exchange interactions in (1) the effective triplet hopping amplitudes a and b in (5) can be of either sign, even for purely AFM $j_{2,4,5,6}$.

It is instructive to link (5) to other analytic results known from related models. In particular, setting $j_1 = j_2$ and $j_{3,5,6} = 0$, the tetrahedral cluster system of fig. 1 is identical to a stack of Heisenberg models on the 1/5-depleted square lattice¹⁹. Bond operator theory (BOT) has been applied to this model yielding a triplet dispersion of $E_T(\mathbf{k}) = [1 + 2(j_4 - 2j_2)/3(\cos(k_x) + \cos(k_y))]^{1/2}$ in the quadrumer phase²⁰. To 1st order this is obviously identical to (5) with the same setting of parameters. Similarly, for $j_{2,3,4,6} = 0$ the quadrumer limit of fig. 1 maps onto the 'dimerized spin-1 chain sector' of the tetrahedral-chain model of the tellurates studied in ref.^{12,13}. BOT has been applied also to that model, leading to $E_T(\mathbf{k}) = [1 - 8j_5/3\cos(k_z)]^{1/2}$. Again, the latter is identical to 1st order with (5) with the same choice of parameters. While this serves as a consistency check for the series expansion we note that BOT, which is approximate only, differs from the exact series already at

2nd order. Additional details on this can be found in the appendix.

To test the quality of our perturbative expansion at 4th order we compare to the plaquette series-expansion of Gelfand and collaborators for the 1/5-depleted square lattice²¹. This is achieved by restricting the parameters in (1) to those of ref.²¹, i.e. $j_1 = \gamma$, $j_2 = \lambda\gamma$, $j_4 = \lambda$, and $j_{3,5,6} = 0$. The plaquette series is a *one* parameter expansion for a 2D model, which allows for expansion of $E_T(\mathbf{k})$ up to 6th order with respect to λ , where the unperturbed Hamiltonian incorporates γ exactly. In fig. 2 we contrast the 4th order results from our *six* parameter expansion for the 3D tetrahedral spin system with the plaquette expansion by considering the triplet dispersion. Despite small deviations which set in upon increasing γ the overall agreement is satisfying. Since γ is treated exactly within the plaquette expansion, our 4th-order quadrumer series does not coincide with one of the edges of the error bars in fig. 2 which refer to the 4th- and 5th-order plaquette series.

Next we analyze the stability of the quadrumer phase against magnetic ordering by identifying the surface in parameter space, closest to $j_{1,\dots,6} = 0$ which allows for triplet softening, i.e. the occurrence of a wave vector \mathbf{k}_C with $E_T(\mathbf{k}_C) = 0$. We emphasize that apart from such instabilities, the tetrahedral spin system may exhibit others transitions, as e.g. those related to the 1st-order, local quadrumer to dimer-product transition on each of the tetrahedra. Here we focus on the triplet softening only. To begin, in fig. 3 we depict the instability surface at 1st order as obtained from (5) along with the critical wave vectors at which softening occurs. Due to the competing interactions several ordering patterns are possible, even for AFM couplings only.

Since the tetrahedral cluster-model contains six exchange coupling parameters we will simplify the stability analysis at 4th order by selecting a subset of them only. This selection is based on the effective exchange constants at 1st order, i.e. we will focus on the stability as a function of a , b , and c setting $j_{1,4,6} = 0$. Figure 4 a) shows the corresponding instability surface. It has been

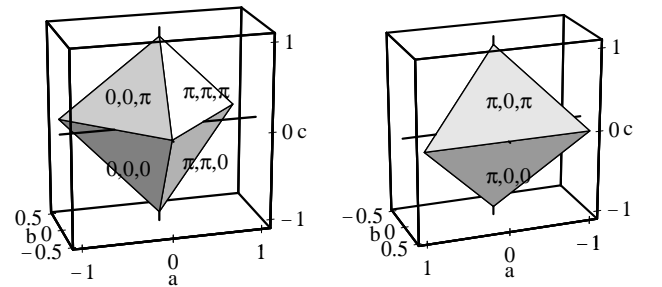


FIG. 3: Left(right) panel: rear(front) view of the stability surface of the quadrumer phase at 1st order. Faces are labeled by the wave vectors \mathbf{k}_C of the instabilities and $a = (j_4 - 2j_2)/3$, $b = j_3/3$, and $c = 4(j_6 - j_5)/3$.

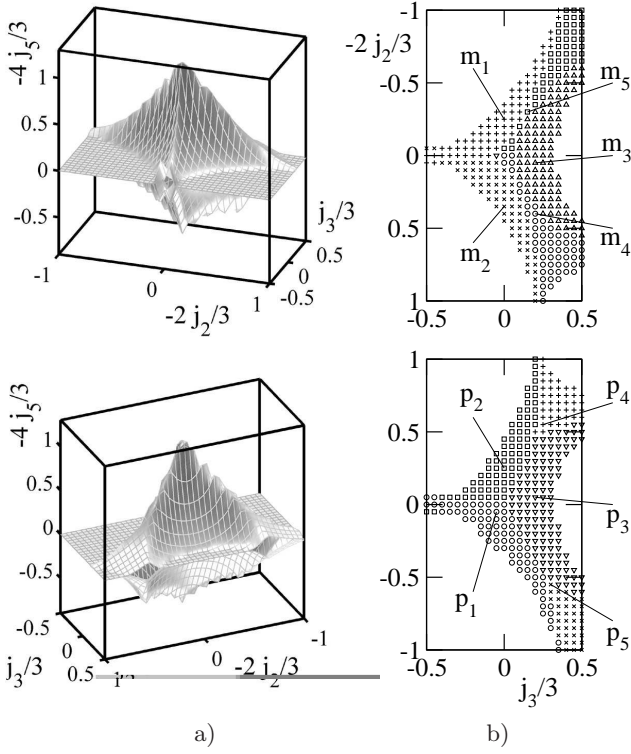


FIG. 4: a) top(bottom) panel: rear(front) view of the stability surface of the quadrumer phase at 4th order. b) top(bottom) panel: wave vectors of instability for $j_5 < (>)0$. Labels refer to \mathbf{k}_C : $+$ $= (0, 0, \pi)$, $\square = (\pi, \pi, 0)$, $\triangle = (\pi, 0, \pi)$, $\nabla = (\pi, 0, 0)$, $\circ = (0, 0, 0)$, and $\times = (\pi, \pi, \pi)$. Selected points $m(p)_{1,\dots,5}$ refer to fig. 5

obtained from a numerical search for zeros of the gap of the 4th-order triplet-dispersion on a mesh of $21 \times 41 \times 2$ points in the abc -space. For this purpose we have used the bare series with no Padé approximations applied. The surface is not closed at its extremal extensions in the ab -plane, rather the stability analysis has been confined to the range of parameters shown in this figure in order to comply with the finite range of convergence of the perturbative result. Only commensurate instability wave-vectors have been found within the range of parameters investigated. The type of these wave vectors is shown in 4 b). While its shape is deformed with a reduced volume, the main features of the 4th-order instability surface are still consistent with those at 1st order. We find that the additional critical wave-vector types with appear along the 'edge-regions', i.e. at $m_{4,5}$ and $p_{4,5}$ occur within a parameter-range of poor convergence of the perturbation theory. Therefore, these may be subject to change at higher orders.

Finally we consider the triplet dispersion at critical coupling strengths. In fig. 5 we show $E_T(\mathbf{k})$ for wave-vectors \mathbf{k} along high-symmetry directions of the Brillouin zone. The exchange parameters have been selected from the points $p_{1,\dots,5}$ and $m_{1,\dots,5}$ on the instability surface of fig. 4 b). The figure demonstrates a rich variety of \mathbf{k} -dependencies possible. Since $\text{Cu}_2\text{Te}_2\text{O}_5\text{Cl}_2$ seems to or-

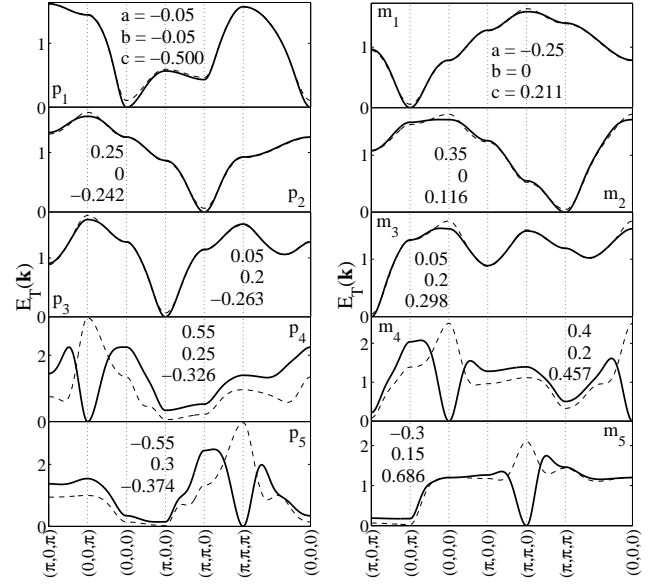


FIG. 5: Left(right) panel: elementary triplet dispersion in the quadrumer phase for $j_5 > (<)0$ at onset of instability. Solid(dashed) line refers to 4th(3rd) order series expansion. x-axis: path denotes path in Brillouin zone. Subplot labels $m(p)_{1,\dots,5}$ indicate location of exchange parameters on instability surface as in fig. 4 b). Insets refer to exchange parameters at $m(p)_{1,\dots,5}$.

der magnetically, inelastic neutron scattering data on the tellurates would be interesting in order to choose among these dispersion for a set of exchange constants relevant to the Chlorine system. To check for the convergence of the series expansion fig. 5 contains both, 3rd- and 4th-order results. On those faces of the instability surface which appear as continuous deformations of the 1st order surface of fig. 3, i.e. for $p(m)_{1,2,3}$ the perturbative result is well converged. However, within the aforementioned edge-regions, i.e. for $p(m)_{4,5}$ the convergence is insufficient. In particular the critical wave-vectors of the instabilities deduced from the 4th-order result within this region may be an artifact. This remains to be clarified in future analysis.

To summarize, we have performed a quadrumer series-expansion for a three-dimensional tetrahedral cluster spin-system using the flow-equation method. We have shown our results to incorporate and interpolate between findings known from previously studied either one- or two-dimensional quantum spin-systems which are found to be limiting subsets of our model. We have analyzed the dispersion of the elementary triplet excitations and the stability of the quadrumer phase against magnetic ordering. Future studies will have to contrast this type of ordering against other transitions possible in this cluster system in order to add more information towards a complete quantum phase-diagram. We hope that our results may prompt further investigations of the tellurate compounds $\text{Cu}_2\text{Te}_2\text{O}_5\text{X}_2$, in particular inelastic neutron scattering studies in order to clarify the relation of the cluster spin-model to these materials.

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APPENDIX A

To further clarify and connect to other existing analytic approaches, in this appendix we also list the result to 2nd order in $j_{1,\dots,6}$ for the triplet dispersion for which we find

$$\begin{aligned}
E_T(\mathbf{k}) = & 1 + \frac{145j_2^2}{432} - \frac{31j_3^2}{864} - \frac{187j_2j_4}{432} - \frac{31j_4^2}{864} + \frac{8j_5^2}{27} \\
& - \frac{52j_5j_6}{27} + \frac{101j_6^2}{108} + \left(\frac{2j_3}{3} + \frac{7j_3^2}{36} - \frac{1}{9}(2j_2 - j_4)^2\right) \\
& \times \cos(k_x) \cos(k_y) + \left(-\frac{j_2^2}{9} + \left(-\frac{1}{3} + \frac{5j_3}{27}\right)(2j_2 - j_4)\right. \\
& \left. + \frac{7j_4^2}{72}\right)(\cos(k_x) + \cos(k_y)) + \frac{1}{54}j_3^2 \cos(2k_x) \cos(2k_y) \\
& + \left(-\frac{j_2^2}{27} - \frac{j_3^2}{18} + \frac{j_2j_4}{27} + \frac{j_4^2}{108}\right)(\cos(2k_x) + \cos(2k_y)) \\
& + \frac{1}{27}j_3(2j_2 - 3j_4)(\cos(2k_x) \cos(k_y) + \cos(k_x) \cos(2k_y)) \\
& + \left(-\frac{2j_5^2}{3} - \frac{4(j_5 - j_6)}{3} + \frac{j_6^2}{9}\right) \cos(k_z) \\
& + \frac{8}{9}j_3(j_5 - j_6) \cos(k_x) \cos(k_y) \cos(k_z) \\
& - \frac{4}{9}(2j_2 - j_4)(j_5 - j_6)(\cos(k_x) + \cos(k_y)) \cos(k_z) \\
& - \frac{4}{9}(j_5 - j_6)^2 \cos(2k_z) .
\end{aligned} \tag{A1}$$

In contrast to the 1st-order result, a dependence on combined effective exchange constants only, i.e. $(2j_2 - j_4)$ and $(j_5 - j_6)$, is absent. For the case of the 1/5-depleted square lattice, i.e. for $j_1 = j_2$ and $j_3 = j_5 = j_6 = 0$, and rewriting (A1) in a form which allows for direct comparison with the BOT of ref.²⁰ we get

$$\begin{aligned}
E_T(\mathbf{k}) = & 1 + \frac{59j_2^2}{144} - \frac{73j_2j_4}{144} - \frac{47j_4^2}{864} \\
& + \left(\frac{1}{3}(j_4 - 2j_2) - \frac{j_2^2}{9} + \frac{7j_4^2}{72}\right)(\cos(k_x) + \cos(k_y)) \\
& - \frac{1}{18}(j_4 - 2j_2)^2(\cos(k_x) + \cos(k_y))^2 \\
& + \left(\frac{4j_2^2}{27} - \frac{4j_2j_4}{27} + \frac{2j_4^2}{27}\right)(\cos(k_x)^2 + \cos(k_y)^2) .
\end{aligned} \tag{A2}$$

this shows the BOT-dispersion, cited after (5), to be correct only to 1st order. Analogous, for the 'dimerized spin-1 chain sector' of the tetrahedral chain studied in refs.^{12,13}, i.e. for $j_{2,3,4,6} = 0$, we may rewrite (A1) into

$$\begin{aligned}
E_T(\mathbf{k}) = & 1 + \frac{20j_5^2}{27} - \left(\frac{4j_5}{3} + \frac{2j_5^2}{3}\right) \cos(k_z) \\
& - \frac{8}{9}j_5^2 \cos(k_z)^2 .
\end{aligned} \tag{A3}$$

Again, the BOT-dispersion is correct to 1st order only. In ref.¹³, perturbation theory up to 2nd order has been performed using a very different method than presented here. Therefore it is satisfying to realize, that (A3) is exactly identical to the corresponding eqn. (9) in section II.B.2. of ref.¹³.

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